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Tritium extraction from lithium-lead in the EU DEMO blanket using Permeator Against Vacuum

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The current studies on the development of the EU DEMO breeding blanket include among the options the use of liquid Lithium-Lead (17Li-83Pb) as tritium breeder (and multiplier), with different coolants. As the tritium is steadily produced in the blanket during the reactor operation, suitably efficient strategies for the tritium extraction system (TES) from the breeder must be developed, allowing a closed fuel cycle in situ and avoiding tritium accumulation in the machine. The Permeator Against Vacuum (PAV) appears to be one of the most promising solutions to achieve this goal. In this paper, the performance of a PAV system is studied by means of different models describing the transport of tritium in the liquid PbLi and in the metallic membrane separating it from the vacuum. The comparison of the results for different membrane materials and size of the device, for a given target efficiency, allows to optimize the PAV design, also taking into account corrosion issues. The approximations and limitations of the adopted models are also addressed.

Keywords: DEMO breeding blanket, Lithium-Lead, Tritium Extraction System (TES), Permeator Against Vacuum (PAV)

I. Introduction

The history of the DEMO breeding blanket designs in the EU is now over 20 years long.[1,2] Several of the options currently under study, namely the Dual-Coolant Lithium-Lead (DCLL), the Helium-Cooled Lithium-Lead (HCLL) and the Water-Cooled Lithium-Lead (WCLL), use liquid Lithium-Lead (17Li-83Pb) as tritium breeder (and multiplier).

As the tritium is steadily produced in the blanket during the reactor operation, and as it is needed to close the fuel cycle in situ, while at the same time it cannot be left to accumulate an inacceptable inventory in the machine, suitably efficient strategies for the tritium extraction system (TES) from the breeder must be developed.

Among the TES options currently under consideration, the permeator against vacuum (PAV), appears to be one of the most promising solution for the DCLL, HCLL and WCLL BB.[3]

The PAV operating principle is simple: PbLi with a certain concentration of tritium flows in a channel delimited for a given length by a membrane permeable to tritium. Vacuum is maintained on the other side of the membrane, so that the difference between the tritium partial pressure on the two sides of the membrane drives the flux of tritium from the PbLi side to the extraction, leaving a lower tritium concentration at the outlet of the channel. As opposed to the undesired permeation of tritium through the walls in the rest of the circuit, which may lead to safety issues and reduced TBR, here the relatively low solubility and the resulting high partial pressure of tritium in PbLi, together with a suitably permeable and corrosion resistant material for the membrane, are at the basis of the PAV potential for operation.

In this paper, two models for the prediction of the PAV efficiency are applied to the HCLL and WCLL. The first model, widely adopted in these kind of analyses, considers only bulk phenomena in the membrane, while the second is proposed to account for surface effects at the interface membrane-vacuum. The models are then used to assess by a sensitivity study how the main TES design parameters affect the PAV system size for a given target efficiency η requested.

II. Model description

In Figure 1 a sketch of the PAV geometry is given, as well as a plot of the radial section of the pipe considered by the models adopted in the following, with the identification of the physical quantities of interest.

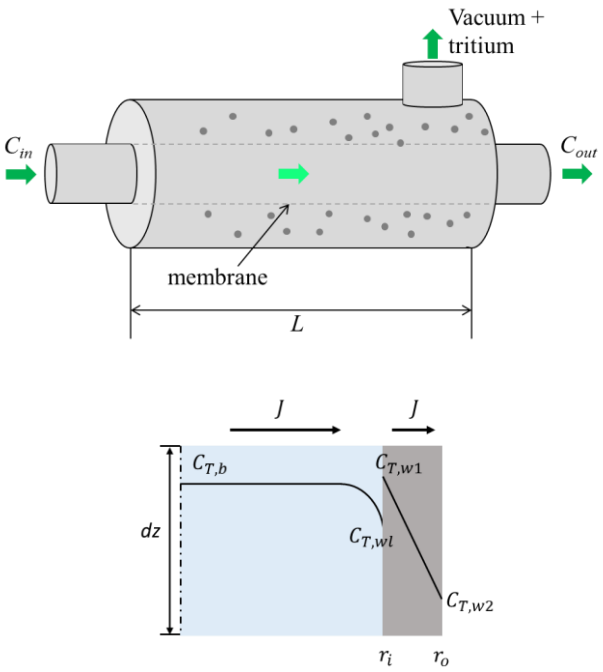

Figure 1: Schematics of the Permeator Against Vacuum (PAV) system (above) and radial section of the channel, with identification of the unknowns of the problem (below).

II.A Simplified analytical model

A first modelling option, adopted in Refs. 4-6 and described in Ref. 5, focuses on the transport of tritium in the PbLi bulk and in the membrane, neglecting the presence of helium bubbles mixed with PbLi and all surface effects on both the PbLi and vacuum side, assuming that permeation through the membrane is influenced only by bulk phenomena (diffusion). As consequence of the latter, in this model the concentration on the vacuum side C_{w2} is assumed to vanish, as a result of the imposition of vacuum (zero

pressure) together with the use of Sievert's law $C = K\sqrt{p}$. At the interface between PbLi and the membrane the continuity of tritium partial pressure is usually assumed in literature, obtaining the following relation through the Sievert constants K_s for the membrane and K_l for PbLi:

$$\frac{C_{T,w1}}{K_s} = \frac{C_{T,wl}}{K_l}. \quad (1)$$

The efficiency is evaluated as $\eta = 1 - C_{out}/C_{in}$. Calculations are carried out requiring a prescribed efficiency of 80% and obtaining the corresponding length of the system L, imposing the other geometrical (radius of pipe and membrane thickness) and physical (temperature and pressure) parameters. Due to the linearity of the model, the efficiency is independent on the inlet concentration C_{in}.

II.B Model accounting for surface effects

While the previous model assumes that permeation through the membrane is influenced only by bulk phenomena (diffusion), a model accounting also for surface phenomena is now introduced. In particular, recombination and desorption processes occurring at the membrane-vacuum interface could be as rate limiting as diffusion, and tritium flux at this interface can be modeled with the relation:[8]

$$J = 2k_R \cdot C_{T,w2}^2 \quad (2)$$

where the recombination constant k_R is introduced. This model no longer requires the imposition of $C_{w2}=0$ on the vacuum side, since Sievert's law is not applied.

No dissociation phenomena occur at the PbLi-membrane interface, since tritium is in atomic form in PbLi, therefore we can assume that the rate of tritium atoms from PbLi to the membrane is in equilibrium condition, thus allowing to use relation (1).

Eq. (2) is coupled with the equations of the previous model[5] in the bulk of PbLi and in the membrane. The result is a set of equations that cannot be solved analytically, and, more importantly, the inlet concentration C_{in} affects the efficiency of the PAV. Since the HCLL and WCLL designs are characterized by different values of C_{in}, this parameter is considered in the parametric analysis presented in the next sections.

III. Results

III.A Comparison of different membrane materials

The first set of results presented focuses on the comparison of the choice of the membrane composition, performed adopting the simplified analytical model. The two main options, based on previous analysis,[6] are iron and niobium. Nb has a higher tritium permeability with respect to Fe, thus allowing to improve the extraction performance, but it tends strongly to oxidize at high temperature. For this reason, a Pd coating should be envisaged, although it is not considered in this preliminary design stage. However, the additional resistance given by the Pd coating should not affect the result significantly because of its high permeability and small thickness.

The data adopted for the comparison are summarized in Table I. Two temperature values are chosen, assuming as minimum the design outlet T of PbLi in the BB, and then considering a higher value, compatible with the material mechanical properties, to increase efficiency. The pipe radius and PbLi velocity are varied in a reasonable range (each curve starts from the minimum velocity which ensures Re=1e4 and therefore a turbulent regime), compatible with manufacturing and corrosion constraints. A parametric analysis on the diffusivity D_l is also performed, since the range of variation of this parameter covers several orders of magnitude.

Table I: Input data adopted in the analytical model calculations for the comparison of Fe and Nb as membrane materials.

η [-]		0.8	
Membrane thickness [mm]		0.5	
T_{PbLi} [°C]		300	550
ρ_{PbLi} [kg/m^3]		9.84e3 (Ref. 9)	9.53e3 (Ref. 9)
μ_{PbLi} [Pa s]		2.15e-3 (Ref. 9)	1.02e-3 (Ref. 9)
D_l [m^2/s]		8.64e-10(Ref. 10)	4.83e-9 (Ref. 10)
		6.51e-10 (Ref. 11)	9.94e-10 (Ref. 11)
K_l [mol/m^3/Pa$^{0.5}$]		2.77e-2 (Ref. 12)	6.28e-2 (Ref. 12)
		9.94e-4 (Ref. 13)	1.05e-3 (Ref. 13)
P_{Nb} [mol/m/s/Pa$^{0.5}$]		6.20e-6 (Ref. 14)	6.45e-7 (Ref. 14)
P_{Fe} [mol/m/s/Pa$^{0.5}$]		1.57e-11 (Ref. 14)	1.46e-10 (Ref. 14)

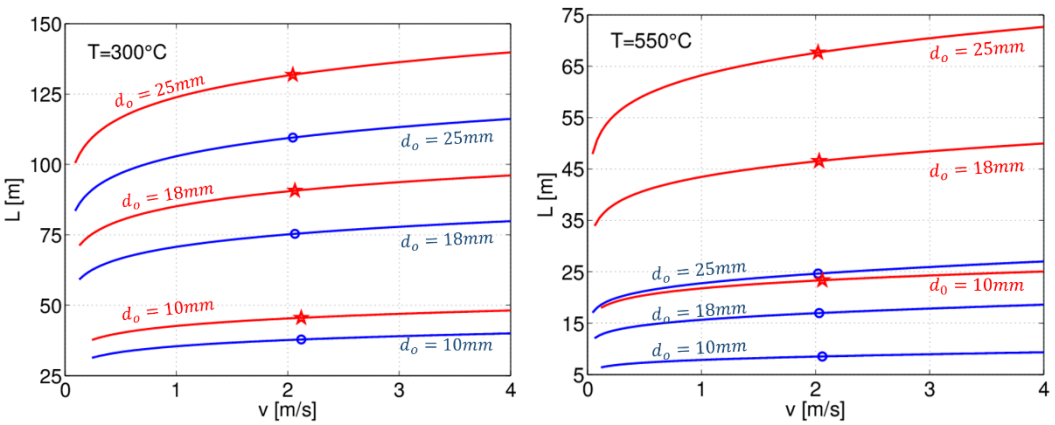

Figure 2: PAV length as a function of PbLi flow velocity adopting as membrane material Nb considering two temperatures of operation and different values of the hydraulic diameter d_o. Circles: D_l value from Ref. 10; stars: D_l value from Ref. 11; K_l from Ref. 12, which is considered the most reliable and conservative value.

The use of a Fe membrane is not feasible (not shown), since very long (from hundreds to thousands of meters, depending on temperature and velocity) channels are needed to achieved the required efficiency. Increasing the diameter affects the results because the ratio between the cross section and wetted perimeter increases. The Nb

length shows a non-linear behavior with respect to velocity, typical when transport across PbLi is rate limiting. PAV shows better performances at higher temperatures, due to the increased PbLi diffusivity.

III.B Comparison of modelling options for the Nb membrane

Having identified Nb as the best option for the membrane material, we now compare the results obtained with the model not accounting for surface phenomena to those obtained with the model accounting for surface effects. To evaluate the permeating tritium flux in this case, the value of k_R in (2) is necessary. k_R can be expressed as:[15]

$$k_R = \sqrt{\frac{2}{3}} \frac{1.3 \cdot 10^{24}}{Na \cdot K_{S0}^2 \cdot \sqrt{T}} \exp\left(\frac{2(E_S - E_C)}{RT}\right) \quad (3)$$

where K_{S0} = 0.127 mol m^{-3} Pa$^{-0.5}$ (Ref. 14) is the pre-exponential coefficient of Sievert constant, E_S = –34 kJ mol^{-1} (Ref. 15) is the heat of solution and E_C = 40 kJ mol^{-1} is the activation energy for dissociative adsorption (considering the more conservative value between 35 and 40 kJ mol^{-1}, both available in ref. 15), *Na* is the Avogadro number. The term $\sqrt{2/3}$ is introduced to take into account that the formula for k_R was originally defined for deuterium.

Since the model depends on the inlet concentration of tritium, we consider the two different values characterizing HCLL and WCLL: 20-30 Pa for HCLL and 50-80 Pa for WCLL. Starting from the inlet pressure of tritium in PbLi, the corresponding concentration C_{in} is obtained applying Sievert's law.

To understand how C_{in} and k_R affect the system performance a parametric analysis is carried out, see Fig. 3. The increase of the inlet concentration in the range 20-80 Pa

leads to a non-negligible reduction of the PAV length (up to 15%), especially if the lower operating temperature is assumed. This is due to the imposed relation (2) between the tritium flux and the concentration, which is affected by its inlet value.

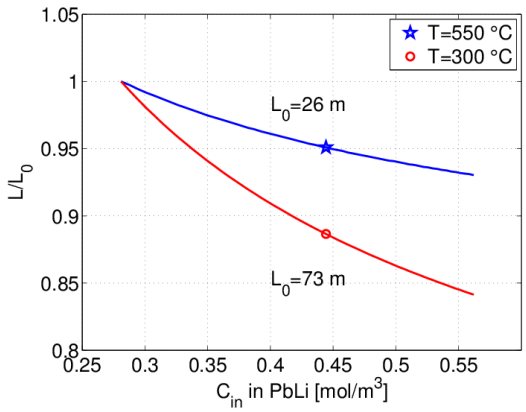

Figure 3: Normalized PAV length as function of inlet concentration. Data adopted: $d_0=10$ mm, $v=0.5$ m/s, K_l from Ref. 12, D_l from Ref. 11 to be conservative.

In Fig. 4 Comparison of the PAV system length as a function of PbLi flow velocity adopting the two modelling options illustrated above.

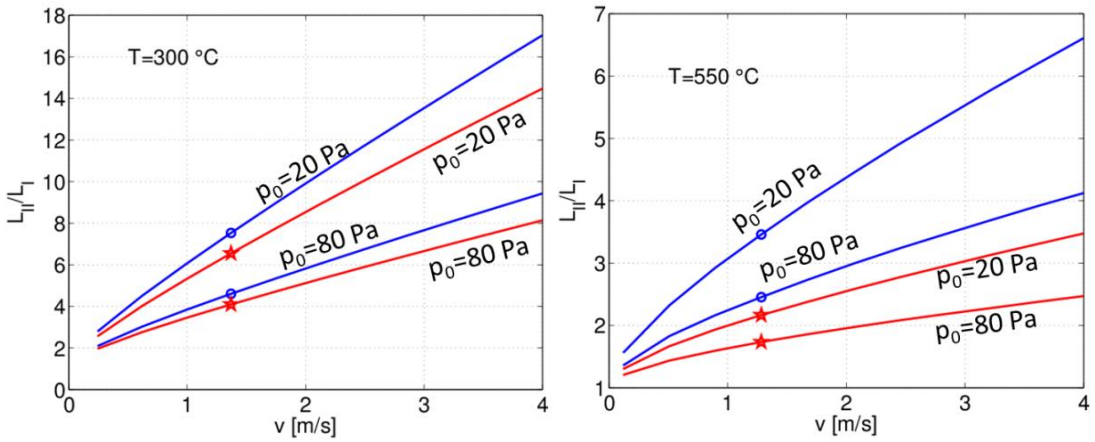

Figure 4: Comparison of the PAV system length as a function of PbLi flow velocity adopting the two modelling options proposed. L_I stands for the length computed with the first model, while L_{II} with the one that accounts for surface effects. Nb membrane assumed, considering two different values of tritium inlet

pressure, two temperatures of operation and d_o = 10 mm. Circles: D_l from Ref. 10; stars: D_l from Ref. 11. K_l from Ref. 12.

Fig.4 demonstrates that surface phenomena play a non-negligible role in tritium transport, affecting the resulting PAV length, which is especially true when the resistance associated to the diffusion in PbLi is lower, i.e. at higher velocities and assuming a higher value of D_l. However, all results show that transport in the membrane is rate limiting and needs to be considered in the system design phase.

The influence of the value of the Sievert constant is also analyzed. The value of K_l affects the solution of the problem since it is used in the evaluation of the inlet condition C_{in} and it also appears in Eq. (1). Of the two roles, the second is prevailing and in fact the use of a smaller Sievert constant (Ref. 13) produces shorter PAV lengths. However, Fig. 5 shows that the uncertainty on K_l strongly affects the PAV length, especially for increasing velocity.

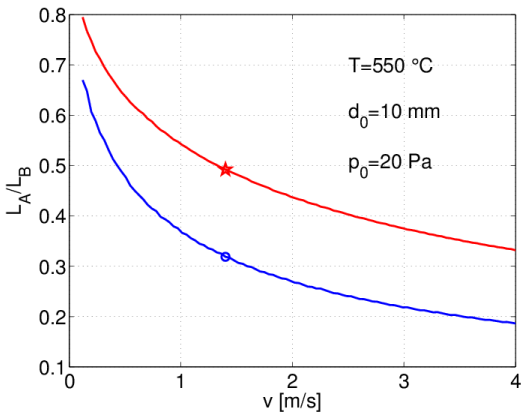

Figure 5: Comparison of the PAV system length with Nb membrane for two different values of Sievert constant. L_A stands for the length computed using K_l from Ref. 13, while L_B with K_l from Ref. 12. Circles: D_l value from Ref. 10; stars: D_l value from Ref. 11.

IV. Conclusions

In this work an analysis of the PAV system is presented, adopting two different models for tritium transport. The first model neglects surface phenomena at material interfaces, assuming diffusion as the limiting phenomenon, while the second takes into account both bulk diffusion and surface phenomena. Both models require a set of parameters characterizing PbLi and the membrane material.

A sensitivity study was performed with respect to the main design parameters (inner PAV channel diameter, PbLi speed and inlet temperature), and input parameters (tritium solubility and diffusivity in PbLi), showing their impact on the PAV length for a prescribed efficiency of 80%.

The results show that the most suitable material for the membrane is Nb, and the comparison of the two modelling options shows that surface phenomena are rate limiting and therefore cannot be neglected. Moreover, both the Sievert constant and the diffusivity, which are known only with high uncertainty, strongly affect the system design.

Both models lead to a value of the channel length of several tens of meters, in order to reach the design extraction efficiency of 80%. Therefore, to limit the geometrical size of the PAV system, the possibility to arrange the pipes in concentric spirals is currently under consideration.

Acknowledgments

The views and opinions expressed herein do not necessarily reflect those of the European Commission.

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